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Abstract

The meteorology and physico-chemical characteristics of aerosol associated with two new cases of long range dust transport affecting western Canada during spring 2006 are described. Each event showed enhancements of both sulfate aerosol and crustal material of Asian origin. However, the events were of quite different character and demonstrate the highly variable nature of such events. The April event was a significant dust event with moderate sulfate enhancement while the May event was a weak dust event with very significant sulfate enhancement. The latter event was interesting in the sense that it was of short duration and was quickly followed by significant enhancement of organic material likely of regional origin. Comparison of these two events with other documented cases extending back to 1993, suggests that all dust events show coincident enhancements of sulfate and crustal aerosol. However, events vary across a wide continuum based on the magnitude of aerosol enhancements and their sulfate to calcium ratios. At one extreme, events are dominated by highly significant crustal enhancements (e.g. the well-documented 1998 and 2001 "dust" events) while at the other are events with some dust transport, but where sulfate enhancements are of very high magnitude (e.g. the 1993 event at Crater Lake and the 15 May 2006 event at Whistler). Other events represent a "mix". It is likely that this variability is a function of the comparative strengths of the dust and anthropogenic SO_2 sources, the transport pathway and in particular the extent to which dust is transported across industrial SO_2 sources, and finally, meteorological and chemical processes.

1 Introduction

During April–May 2006, INTEX-B (the Intercontinental Chemical Transport Experiment) was focused on the North Pacific with the goal of providing detailed chemical analysis of tropospheric air following a trans-Pacific pathway to the North American continent. This intensive field campaign, involving ground based, airborne and satellite observations,

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was prompted by over a decade of observational and modelling studies demonstrating the relatively rapid (especially during springtime) trans-Pacific transport of both crustal material (predominately desert dusts) and anthropogenic pollutants from sources in Eurasia to North America (e.g. Jaffe et al., 1999, 2003; Husar et al., 2001; Holzer et al., 2003; Jacob et al., 1999). In this context, the mobilisation and transport of mineral dust from the arid regions of the world is considered of major significance due to the role airborne crustal material plays in the global radiation balance (and hence climate forcing), cloud processes, atmospheric chemistry, oceanic and terrestrial biogeochemical processes (e.g. dust is a major source of iron and calcium), as vectors for microbes, and as a factor influencing local air quality in both "source" and "sink" regions (Prospero et al., 2002). Furthermore, recent studies suggest that dust participates in important interactions with anthropogenic pollutants such as sulfur dioxide (SO_2 – a source of sulfate aerosol). Such pollutants are often mixed into, and interact with, the dust plumes as they pass over urban/industrial sources (Li-Jones and Prospero, 1998; Heald et al., 2006).

Studies of trans-Pacific dust transport suggest a strong linkage between anthropogenic sulfate aerosol and crustal dust transport from Asia. For example, VanCuren's (2003) analysis of aerosol data at Crater Lake, (Oregon) and Mt. Lassen (California) shows that a mixture of dust and combustion products dominate Asian aerosol that arrives in North America. Major fine particle (<2.5 μm diameter) constituent fractions are 30% mineral, 28% organic compounds, 4% elemental carbon, 10% sulfate, <5% nitrate, and <1% sea salt. Heald et al. (2006), using satellite imagery, GEOS-Chem model and Interagency Monitoring for Protected Visual Environments (IMPROVE) surface network data from the Western USA, have also demonstrated the strong coincidence of crustal dust and sulfate aerosol in trans-Pacific pollutant transport. Springtime Asian sulfate aerosol enhancements are shown to be greatest in Washington State and southern British Columbia with maximum 24-h enhancements reaching approximately 1.5 μgm^{-3}. These results suggest that combined dust/sulfate transport events may have important implications for the setting and attainment of visibility standards

in western North America. This is particularly relevant when set against a background of burgeoning emissions of SO_2 from Asia, where not only have SO_2 emissions increased by 119% between 1980–2003 (Ohara et al., 2007), but also Asian sulfate influx to North America has increased 2.4–3.4 fold between 1985–2006 (van Donkelaar et al., 2008). Streets and Waldhoff (2000) indicate that Asian SO_2 emissions are projected to increase from 25.2 mt in 1995 to 30.6 mt in 2020 (assuming emission controls are implemented on large power plants) and possibly to 60.7 mt without emission controls.

Beginning with the well-documented 1998 dust episode, a growing inventory of trans-Pacific dust events affecting North America has been established. Most of these events have involved sources in the Gobi and Takla Makan deserts Asia (Husar et al., 2001; Jaffe et al., 2003; Thulasiraman, 2002), although recently, a case of Saharan dust transport across Asia and the Pacific to North America has been documented (McKendry et al., 2007). In addition, modelling studies have identified many aspects of the climatology, inter-annual variability and pathways of dust transport and deposition (Holzer et al., 2003, 2005; Gong et al., 2006; Zhao et al., 2006). The meteorological mechanisms associated with boundary layer – free tropospheric (BL-FT) exchange of dust (and anthropogenic pollutants) have also largely been identified. BL-FT exchange processes most relevant to dust source areas include the warm conveyer belt (WCB) mechanism (Cooper et al., 2004; Stohl et al., 2002) and frontal lifting, while in receptor regions (such as western North America) subsidence, and mountain wave activity are important in bringing mid- tropospheric dust layers in range of BL entrainment processes (McKendry et al., 2001; Hacker et al., 2001).

Against this background of improved understanding of many aspects of trans-Pacific dust transport, the intensive observations afforded by INTEX-B provide a further opportunity to extend the inventory of documented dust events and to examine the commonalities emerging from such a catalog of events. In this context, our goals in this paper are to:

– Describe two separate dust events identified during INTEX B.

– Compare and contrast these events with previously documented cases, particularly with respect to their magnitude and chemical signature. For the latter, sulfate aerosol measurements during INTEX-B provide a rare opportunity to examine the linkage between sulfate and mineral aerosol in these cases.

2 Methods

Measurements of particles and trace gases are made by Environment Canada at a high elevation site in Whistler, BC, approximately 100 km north of Vancouver (Fig. 1). The site is located at the top of Whistler Peak (2182 m above sea level). There are no continuous combustion sources at the peak and influences from snowmobiles have been identified and removed from the data set (Macdonald et al., 2006). During the period of interest, particle size distributions were measured both with a TSI Scanning Mobility Particle Sizer (SMPS) Model 3934 and a Grimm optical particle counter (OPC) (Model 1.108). The SMPS was used to measure mobility diameter in the 0.01 μm to 0.4 μm size range while the OPC provided optical diameter from 0.3 μm to 20 μm. Particle Chemistry is from bulk filter packs, collected every 48 h and analyzed for inorganic ions by ion chromatography. All particle sampling takes place through a heated stainless steel manifold and filter packs are preceded by a cyclone operated at approximately a 2 μm size cut. Particle sampling is suspended in the presence of fog to ensure no contamination of samples by cloud and that the sampling inlet remains dry and free of rime. In addition, A high mass resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was deployed at the summit of the Whistler Mountain from 20 April to 17 May 2006 (Sun et al., 2008)[1].

[1]Sun, Y., Zhang, Q., Leaitch, R. W., Macdonald, A. M., Hayden, K., Li, S.-M., Liggio, J., Liu, P. S. K., Cubison, M., Worsnop, D., van Donkelaar, A., and Martin, R. V.: Size-Resolved Aerosol Chemistry at Whistler Summit, Southwestern Canada with a High-Resolution Aerosol Mass Spectrometer, J. Geophys, Res., in preparation, 2008.

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Measurements of O_3 (TECO 49), CO (Aerolaser), size distributions of aerosol particles (CPC, PCASP100X, FSSP300) and the chemical size distribution (Quad-AMS) were conducted from a Cessna 207. Flights were approximately two hours long, and most flights consisted of ascent and descent profiles at Whistler to an altitude of about 5.3 km. Data were averaged every second, except for the AMS that was averaged at one minute intervals in order to try to optimize temporal resolution and sufficient signal for detection. Further details of the instrumentation and comparisons are discussed by Leaitch et al. (2008)[2].

Back-trajectories from Whistler were calculated using the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (www.arl.noaa.gov/ready/hysplit4.html). The model is the newest version of a complete online system for computing simple air parcel trajectories to complex dispersion and deposition simulations for any location and date (depending on data availability) using a variety of standard data input products (e.g. the NCEP Reanalysis 1948–present). Further details and validation of the model can be found in Draxler and Hess (1998).

3 Results

3.1 Dust events at Whistler

Observations at Whistler Peak revealed at least four distinct aerosol episodes during April–May 2006 (Fig. 2). This is consistent with observations of sand/dust storm events (SDS) in Northeast Asia (Zhou et al., 2007), Aeronet observations from Saturna Island (Fig. 1) and modeling results described by Zhao et al. (2007). Of these episodes, two are particularly noteworthy in the context of this study. The first, of highest magnitude,

[2]Leaitch, W. R., Macdonald, A. M., Anlauf, K. G., Liu, P. S. K., Toom-Sauntry, D., Li, S.-M., Liggio, J., Hayden, K., Wasey, M. A., van Donkelaar, A., Duck, T., Martin, R., Zhang, Q., Sun, Y., McKendry, I., and Cubison, M.: Vertical profiles of aerosols and ozone at Whistler, BC during Intex-B, in preparation, 2008.

occurred from 23–26 April at Whistler (Fig. 2) and was associated with material with modal particle diameter in the range 2–3 μm. The second occurred from 15–16 May (Fig. 2), was of somewhat lesser magnitude and was notable in the sense that there appeared to be a shift late in the event to a particle size distribution dominated by fine particles (~0.2 μm). Both events were in broad agreement in terms of magnitude and timing with NAAPS forecasts for Cheeka Peak (available at www.nrlmry.navy.mil/aerosol/), a nearby atmospheric chemistry monitoring site.

3.2 Meteorology and Asian dust storms

GEOS-Chem v7-04-09 at 2°×2.5° (www.as.harvard.edu/chemistry/trop/geos/) was used to estimate dust emissions and transport from Asia to North America. It is driven using assimilated meteorological data from the Goddard Earth Observing System (GEOS-4) at the NASA Global Modeling Assimilation Office (GMAO). The dust module is described in detail by Fairlie et al. (2007) and includes the effects of gravitational settling as well as wet and dry deposition.

The top panel of Fig. 3 shows simulated daily mean dust emissions over China between 6 April to 19 May 2006. The bottom panel plots simulated total daily dust emissions within enhancement regions, as outlined within the blue and red boxes, and corresponding to the Taklamakan and Gobi deserts, respectively. Significant dust emissions are produced on 16 April and 7–8 May, and present a likely origin for the dust events observed at Whistler Peak on 23–26 April and 15–16 May. Zhou et al. (2007), in an inventory of SDS activity in 2005 and 2006, also note severe SDS activity in the period 15–19 April 2006 in Mongolia that influenced regions including northern China, the Korean Peninsula and Japan.

Figure 4 shows the simulated transport across the Pacific Ocean of dust aerosol generated from the 16 April and 8 May emission events. To concisely represent plume flow, this figure is generated by regularly sampling the simulated aerosol optical depth of dust at constant longitudinal spacing from the emission date until arrival at Whistler Peak. As in previously documented dust incursions (Husar et al., 2001; Jaffe et al.,

2003; McKendry et al., 2007) both events involved direct transport across the Pacific in well developed zonal flow. Both cases took approximately a week for transport. Columnar flux patterns, shown as blue arrows on Fig. 4, suggest two distinct pathways taken during transport, with the April event arriving at Whistler Peak via a northerly route and the May event from the south. This may be an important factor contributing to differences in the chemical signatures of the two events (see below).

3.3 Whistler observations: vertical profiles and lidar

Vertical profiles of particle counts, and sulfate and organic aerosol concentrations derived from Cessna flights on 25 April and 15 May are shown in Fig. 5a–c. The April 25 fine particle aerosol profile (Fig. 5a) is marked by two distinct components. In the boundary-layer (BL), which extended to about 3 km during late afternoon, the particles were composed of about 60% organic material, 30% sulfate, and much of the balance was made up by nitrate and ammonium. Above 3 km, the fine particle aerosol was almost completely dominated by sulfate. The number concentrations of particles $<1\,\mu$m (i.e. PCASP and 7610) were higher in the BL and much lower above 3 km, consistent with the transport of dust from Asia. The presence of a predominantly sulfate aerosol is also consistent with past observations (e.g. Brock et al., 2004) and other observations during Intex-B (e.g. Peltier et al., 2008; Dunlea et al., 2008[3]; van Donkelaar et al., 2008), and suggest that the aerosol was formed by the oxidation of SO_2 during transport, in both cloud and the gas phase. The fine particles above 3 km were also larger than those in the boundary layer. This is evident in the PCASP size distributions (not shown) and from Fig. 7; the reduction in particle number concentrations from the BL to above 3 km is about a factor of four whereas the reduction in the fine particle mass is a factor of two or less. An increase in the size of the fine particles is an indication of

[3]Dunlea, E., DeCarlo, P. F., Kimmel, J. R., Aiken, A. C., Peltier, R., Weber, R., Tomlison, J., Collins, D., Shinozuka, Y., Howell, S., Clarke, A., Emmons, L., Apel, E., Pfister, G., van Donkelaar, A., Millet, D., Heald, C., and Jimenez, J.-L.: Evolution of Asian aerosols during transpacific transport in INTEX-B, Atmos. Chem. Phys. Discuss., in preparation, 2008.

a longer lived aerosol. Ozone is relatively high in the BL, and it increases from about 65 ppbv at 3 km to about 70 ppbv at 4.7 km. It increases another 10 ppbv in the plume between 4.7 and 5.2 km, evidence that photochemical production of ozone accompanied the production of sulfate. The INTEX-B observations were interpreted with a global chemical transport model (GEOS-Chem) to estimate that Asian anthropogenic emissions during the period increased the mean profiles observed over Whistler for fine particle sulfate by 0.3–0.5 μgm^{-3} (van Donkelaar et al., 2008) and for ozone by 6–8 ppbV (Walker et al., 2008[4]).

As discussed by Zhang et al. (2008), a predominantly sulfate plume impacted the Peak site on the morning of 15 May (Fig. 5b). The profile data show an increase in sulfate between 2 km and 2.5 km at an elevation coincident with Whistler Peak. Accompanying this was a modest increase in O_3 and a substantial increase in CO (Leaitch et al., 2008).

Lidar imagery from the morning of 15 May 2006 (Fig. 6) confirms the presence of an aerosol layer at approximately the elevation of the Peak station (2182 m a.s.l.) and is in agreement with the profile data at this time (Fig. 5b). During the course of the May 15 event, lidar suggests that subsidence was a significant process bringing the sulfate-rich aerosol layer to mountain ridge level. The importance of subsidence in bringing dust layers in reach of the planetary boundary-layer over mountainous regions of western North America was identified by Hacker et al. (2001).

The situation changed dramatically by the afternoon of 15 May (Fig. 5c). While there was still a thin sulfate plume similar to that during the morning, the fine particle aerosol was clearly dominated by organics between 2 km up to about 4.4 km. This abrupt change was also seen in the Peak data (Fig. 7, case 2). Trajectories

[4]Walker, T. W., Martin, R. V., van Donkelaar, A., Leaitch W. R., Macdonald, A. M., Anlauf, K., Cohen, R. C., Huey, G., Avery, M., Weinheimer, A., Flocke, F., Tarasick, D., Thompson, A., Streets, D. G., Ziemke, J., Bucsela, E., and Celarier, E.: Trans-Pacific transport of reactive nitrogen and ozone to Canada during spring, Atmos. Chem. Phys. Discuss., in preparation, 2008.

for this time indicate subsiding air at the 2–5 km level, with origin from the south to south west over the previous one to two days (Fig. 8). Longer range trajectories come from the mid-Pacific and northern China (Fig. 7). Sun et al. (2008) attribute the origin of the organic plume (Case 2, Fig. 7) to regional sources with likely contributions from the urbanized regions extending from southern Puget Sound to Georgia Strait (Seattle – Bellingham-Vancouver) with possible contributions from biomass burning that was evident along the California-Oregon border during the period (map.ngdc.noaa.gov/website/firedetects/viewer.htm). In addition, given elevated tree emissions in the Whistler valley during the time period, biogenic secondary organic aerosol might also have contributed to the observed enhancement of organic aerosol mass.

3.4 Comparison with previously documented cases

In order to compare the dust events observed at Whistler during April–May 2006 with other documented dust events, IMPROVE data from the closest high altitude sites, with a sufficiently long record to encompass the Spring 1993 event, were examined (Crater Lake, and Mt Hood, Oregon). Crater Lake is considered to be a pristine high altitude site that has been used in previous studies to examine trans-Pacific transport (Jaffe et al., 2005; Van Curen and Cahill, 2002; Zhao et al., 2007) and at approximately 2000 m, it is of similar elevation to Whistler. Furthermore, Ca (an important element of the crustal dust signature along with Si, Fe and Al) and SO_4 concentrations measured as part of fine aerosol IMPROVE monitoring are comparable to the filter pack measurements made at Whistler.

Documented trans-Pacific dust events (including those of 2006) are listed in Table 1, while the time series of Ca and SO_4 at Crater Lake are shown in Fig. 9a, b respectively. At Whistler, Ca concentrations reached 0.14 μgm^{-3} on 24 April 2006 and were the highest since the start of record in 2002. Concentrations during the May event were approximately half this amount. Sulfate concentrations at Whistler for the 23–26 April and 15 May events reached 1.1 and 1.8 μgm^{-3} respectively. When placed in the

context of the longer term record from Crater Lake (Fig. 9), the dust events pale in comparison to the large events of 1998 and 2000 when calcium concentrations were enhanced by factors of 30–40 over mean background levels. However, the dust event of 23–26 April 2006 observed at Whistler (and apparent at Crater Lake) appears to be one of the largest 4–5 events observed over the thirteen year period. The May 2006 dust event on the other hand, is of low magnitude with concentrations comparable to the Saharan dust event observed in 2005 (McKendry et al., 2007).

For all events listed in Table 1, aerosol sulfate concentrations at Crater Lake were enhanced over annual mean background concentrations by factors in the range 2–5.6. This is smaller than the range of enhancements shown for crustal dust and likely reflects the relatively constant anthropogenic source strength for Asian SO_2 compared to the highly variable dust sources. Of particular note is the large magnitude 15 May 2006 sulfate aerosol event that produced concentrations at Whistler peak ($1.8\,\mu gm^{-3}$) exceeding the maximum sulfate aerosol associated with documented crustal dust events observed at Crater Lake ($1.74\,\mu gm^{-3}$ on 28 April, 1993).

Ratios of SO_4 to Ca fine aerosol at Crater lake (Table 1) indicate some measure of the "mix" of aerosol for the different events, and together with enhancement factors, provide the basis for a somewhat subjective characterization of "events" with respect to the relative mix and magnitude of the sulfate vs mineral dust components (column two of Table 1). On this basis it is possible to define major dust events (29 April, 1998, 16 April 2001, 23–26 April 2006) in which there was low to moderate sulfate enhancement. In other cases, the event incorporated dust but was a significant sulfate event (28 April 1993, 15 May 2006 at Whistler). Other cases (16–24 March 1999, and the Saharan dust event of 2005) are clearly of a more mixed nature with neither the sulfate or dust enhancements being particularly pronounced.

4 Discussion and conclusions

During INTEX B, two significant aerosol events incorporating Asian dust were observed at Whistler Peak and in nearby aircraft profiles over southwestern British Columbia. Both events showed enhancements of both sulfate aerosol and crustal material of Asian origin. However, the events were of quite different character and demonstrate the highly variable nature of such events. The April event was a significant dust event with moderate sulfate enhancement while the May event was a weak dust event with very significant sulfate enhancement. The latter event was interesting in the sense that it was of short duration and was quickly followed by significant enhancement of organic material likely of regional origin.

In terms of the meteorology of the two dust events observed during INTEX B, both were traceable to typical spring dust storms in China and shared the common pattern evident in previously documented cases (e.g. Husar et al., 2001; McKendry et al., 2005) of subsidence over western North America. Trajectories in the eastern Pacific were quite different in the two cases and were likely responsible for the differences in chemical "signature" of the two events. In contrast to the northerly approach to western North America in the April case, the May 2006 episode trajectory approached British Columbia from the south. This particular trajectory was likely responsible for the switch to high organic concentrations late on the 15 May, likely a response to regional sources over western North America and to the south of Whistler.

In summarizing the documented Asian dust events in Western North America, it is apparent that, in agreement with Heald et al. (2006), all dust events appear to show coincident enhancements of sulfate and crustal aerosol. However, events appear to vary across a wide continuum based on the magnitude of aerosol enhancements and their ratios. At one endpoint are events dominated by highly significant crustal enhancements (e.g. the well documented 1998 and 2001 "dust" events) while at the other end of the spectrum are events with some dust transport, but where sulfate enhancements are of very high magnitude (e.g. the 1993 event at Crater Lake and the 15 May 2006

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event at Whistler). Other events lie somewhere between. It is likely that this variability is a function of:

1. the comparative strengths of the dust and anthropogenic SO_2 sources;
2. the transport pathway and in particular the extent to which dust is transported across industrial SO_2 sources
3. meteorological and chemical processes.

The coincident transport of sulfate and mineral aerosol evident in these events has potential implications for air quality compliance and visibility in western North America, and is likely evolving due to changes in land-use and anthropogenic emissions in Eurasian source regions. Furthermore, transport of sulfate and dust from Asia may have significant impacts on climate due to changes in particle size distributions and increased residence times of particles in the atmosphere.

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Table 1. Chemical Comparison of Documented Trans-Pacific Dust Events, 1993–2006.

Date of West Coast Arrival	Character of Dust/sulfate mix	Crater Lake (1996 m) Values in brackets are from Whistler – 2100 m elevation μgm^{-3}			Enhancement Factor (Compared to overall mean) μgm^{-3}		Publications
		Ca	SO_4	SO_4/Ca ratio	Ca	SO_4	
28 April 1993	Moderate dust/high sulfate	0.12	1.74	14.5	6.2	5.6	Jaffe et al. (2003)
29 April 1998	High dust/moderate sulfate	0.82	1.31	1.6	41.1	4.2	Husar et al. (2001)
16–24 March 1999	Low dust/low sulfate	0.06	0.69	11.5	3.0	2.0	NRL Case Study
16 April 2001	High dust/low sulfate	Missing 0.40*	1.18 0.95*	2.4*	−31*	3.8 2.4*	Jaffe et al. (2003); Thulasiraman (2002); Heald et al. (2006)
15 March 2005	Moderate Saharan Dust/low sulfate	0.10 (0.05)	0.68 (0.70)	6.8	5.1	2.2	McKendry et al. (2007)
23–26 April 2006	High dust/low sulfate	0.20 (0.14)	0.93 (1.1)	4.7	9.9	3.0	this work, van Donkelaar et al. (2008); McDonald et al. (2008); Leaitch et al. (2008); Zhang et al. (2008)
15 May 2006	Low dust/high sulfate (Whistler)	0.05 (0.08)	0.81 (1.8)	16.2	2.6	2.6	this work; van Donkelaar et al. (2008); McDonald et al. (2008), Leaitch et al. (2008); Zhang et al. (2008)

*Mt Hood Oregon (1531 m) values

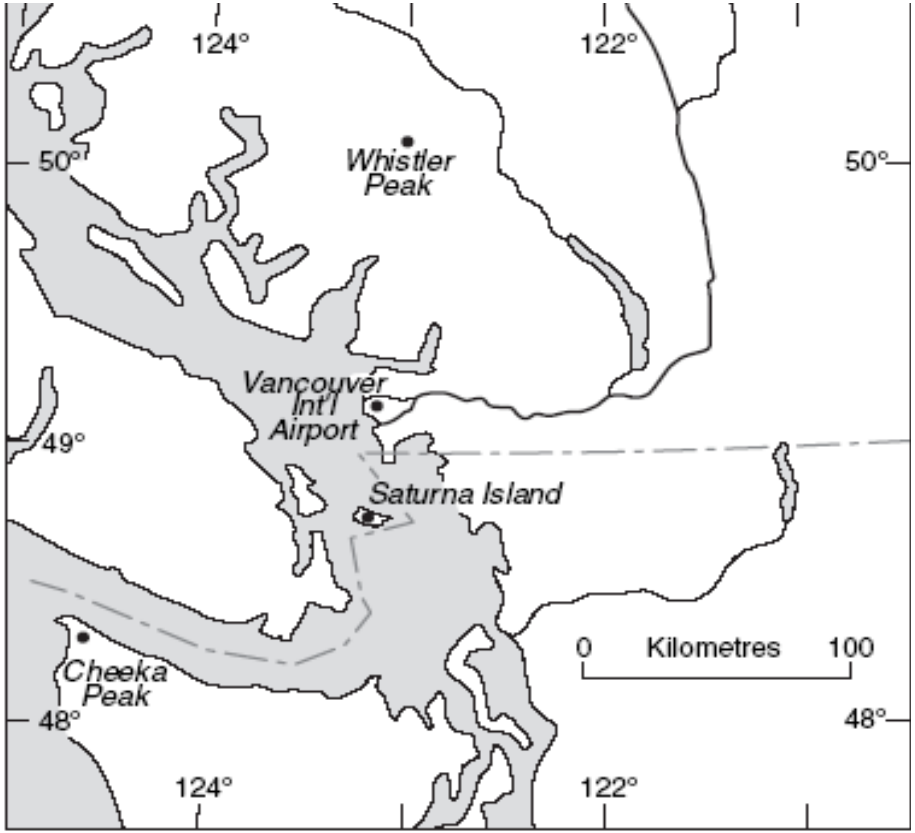

Fig. 1. Map showing study area and places mentioned in the text.

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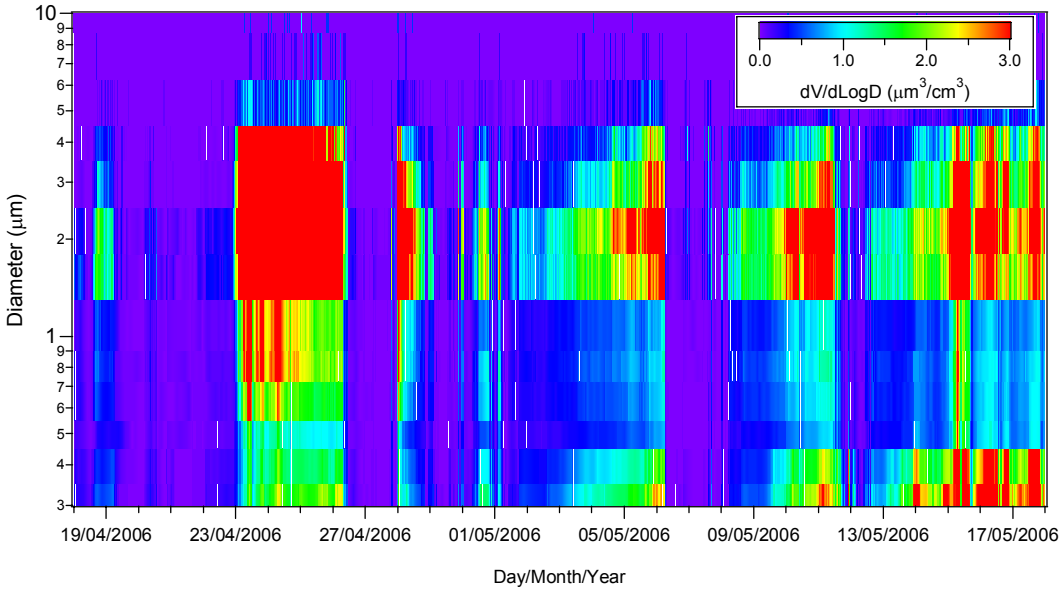

Fig. 2. Two-dimensional image plot from the SMPS at Whistler Peak. Vertical axis is particle diameter (μm).

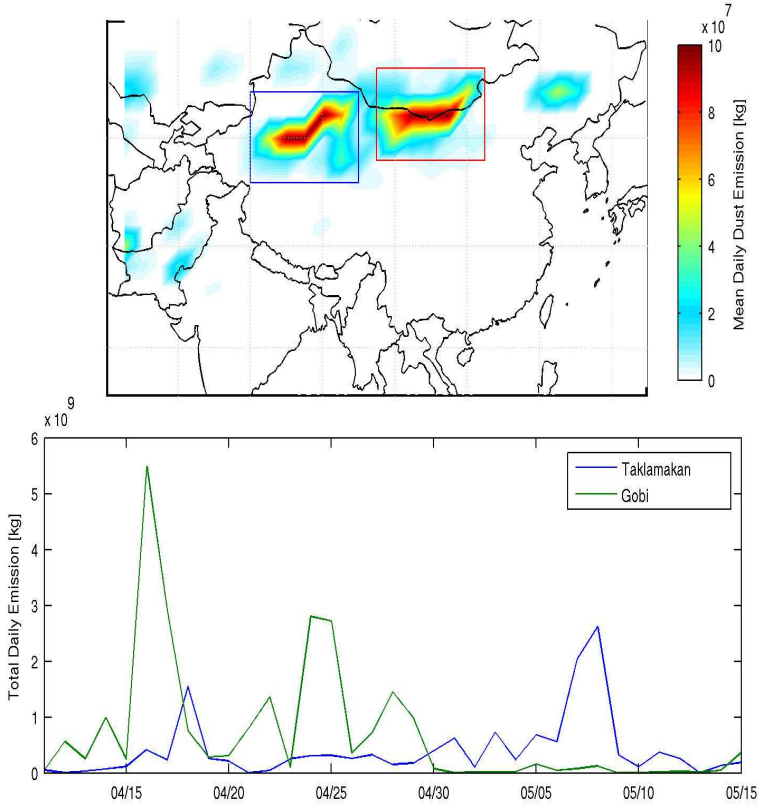

Fig. 3. Simulated dust emissions over East Asia during INTEX-B. The top panel shows average emissions for this period. The bottom panel shows temporal variation of total daily emissions within the regions outlined in the upper panel.

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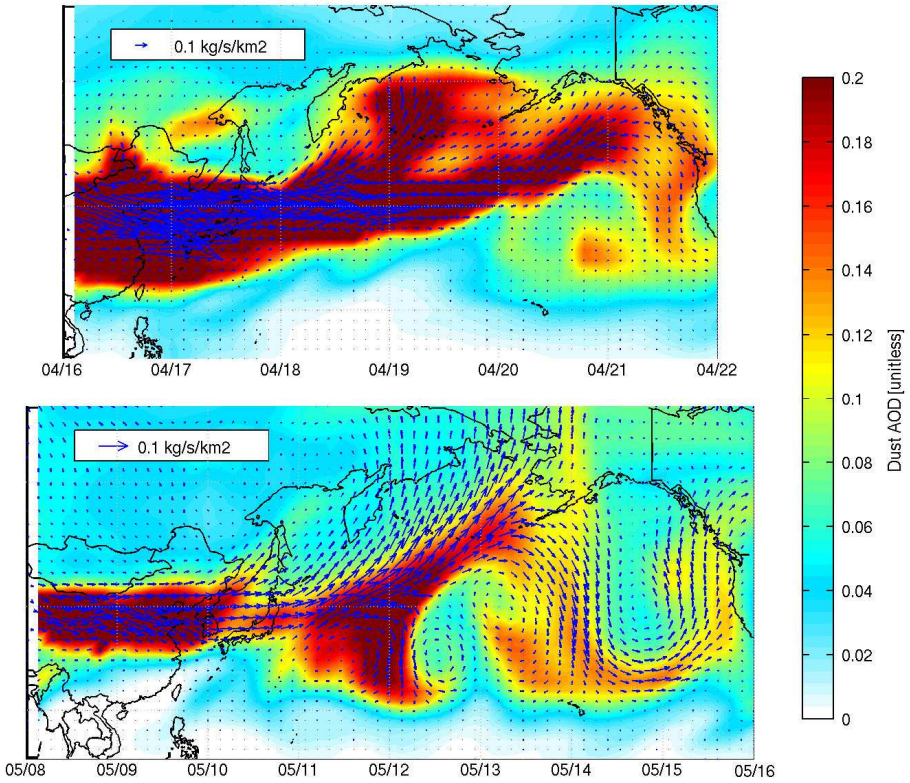

Fig. 4. Transport pathways of East Asian dust plumes. Both panels show the simulated dust component of aerosol optical depth sampled at the dates shown, approximately following plume flow between simulated emission and observation at Whistler Peak. Arrows denote columnar flux magnitude and direction.

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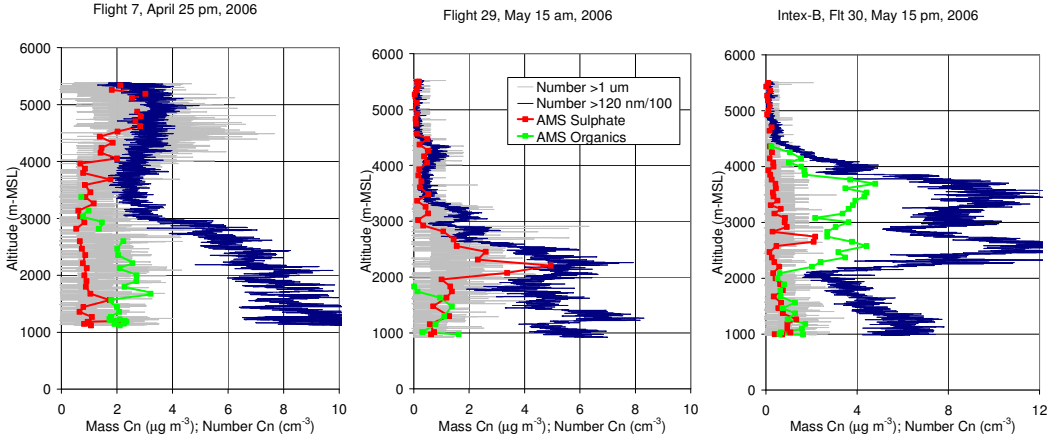

Fig. 5. Vertical profile data of sulfate and total organic fine particle mass concentrations (measured with a quadrapole AMS) and the number concentrations of aerosol particles larger than 120 nm diameter and larger than 1 μm diameter.

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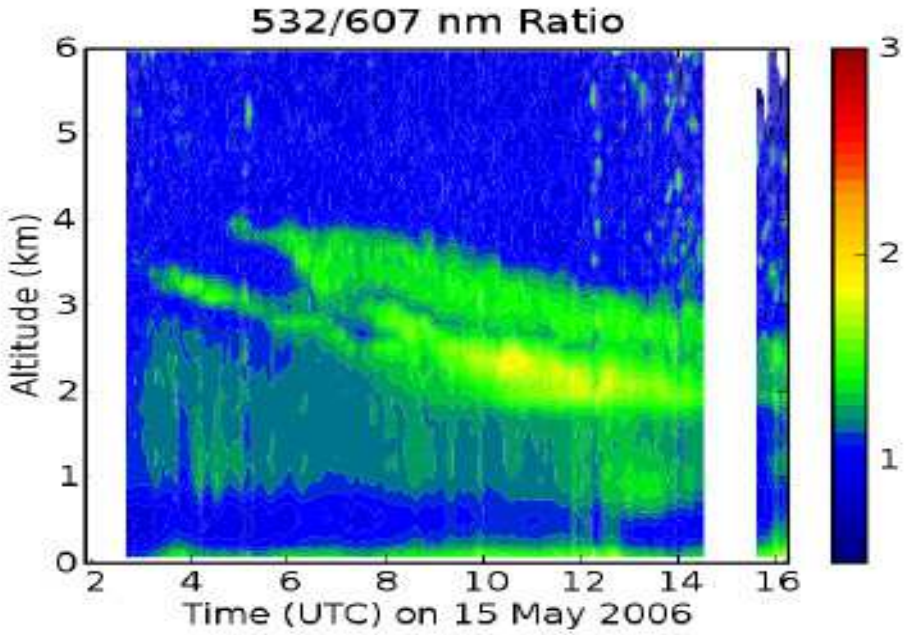

Fig. 6. Lidar imagery showing subsiding aerosol layer (Green) at Whistler on 15 May 2006.

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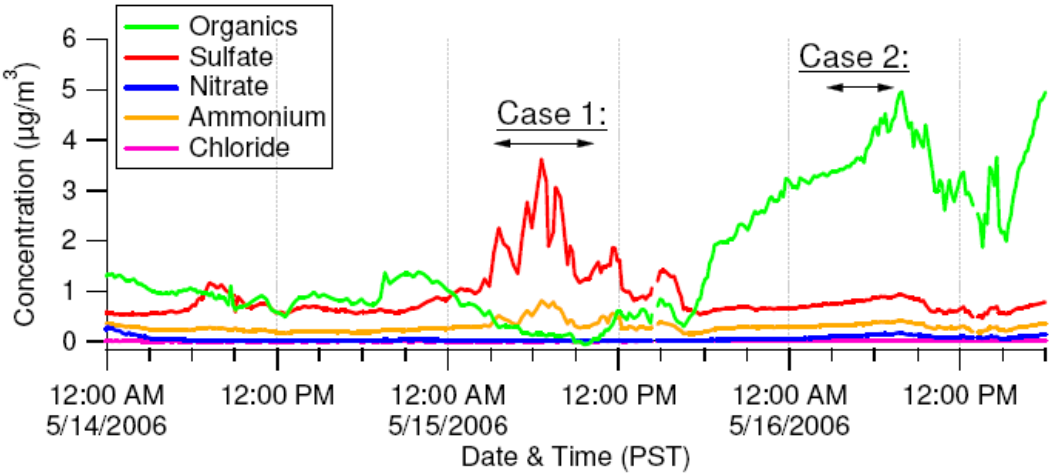

Fig. 7. Time series of the mass concentrations of total organic, sulfate, nitrate, ammonium and chloride in the fine particles as measured at Whistler Peak using a high-resolution time-of-flight Aerosol Mass Spectrometer (W-TOF AMS). From Zhang et al. (2008).

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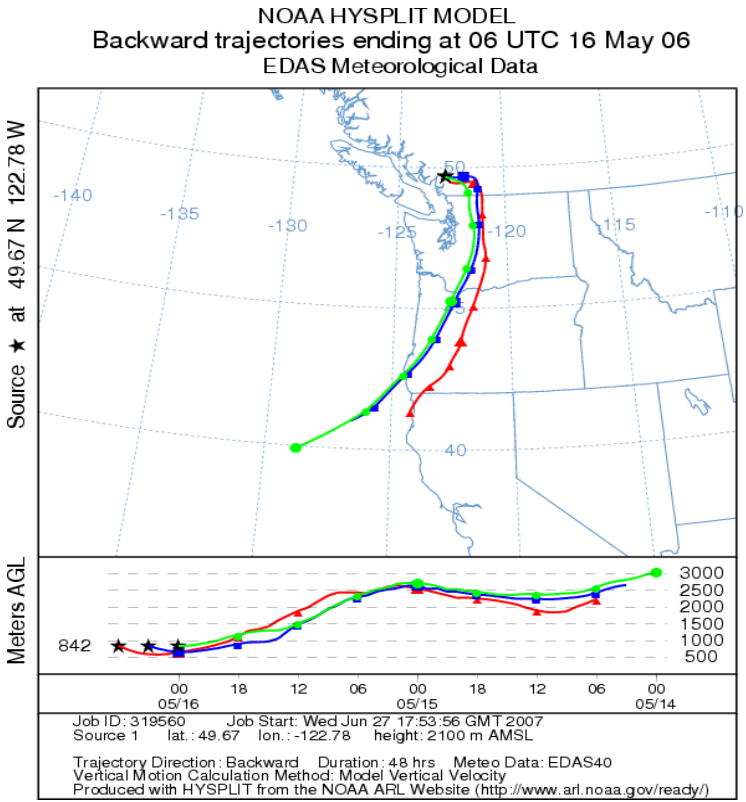

Fig. 8. Back-trajectories from Whistler calculated by HYSPLIT ending in the period 16:00–24:00 LST on 15 May 2006 (i.e. CASE 2 in Fig. 7).

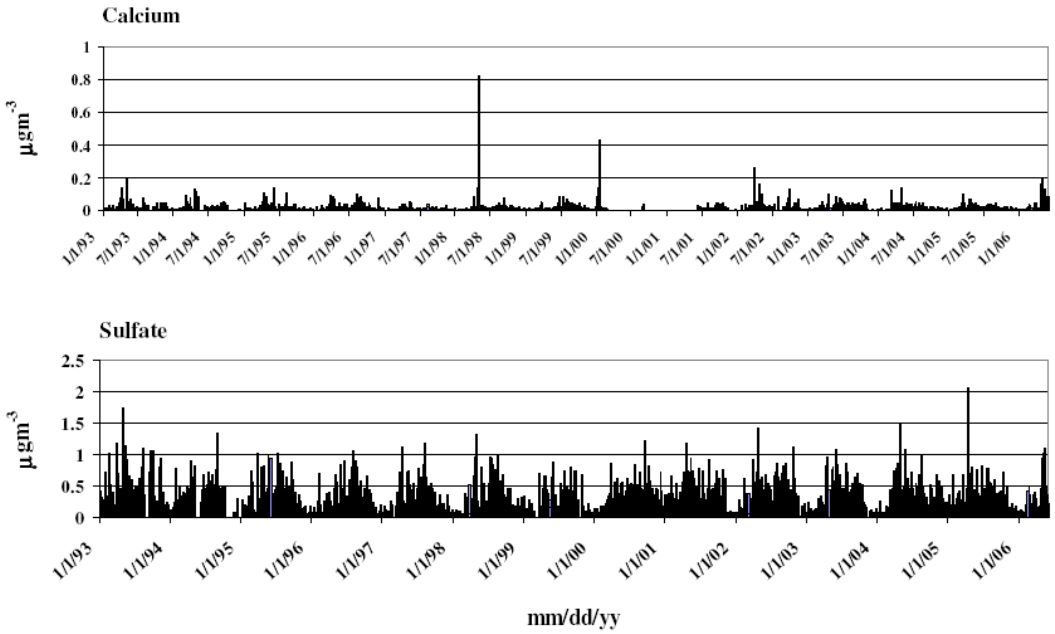

Fig. 9. Elemental Calcium and Sulfate aerosol concentrations measured at Crater Lake Oregon (IMPROVE) 1993–2006.

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